Airborne Flux Observations Constrain Sources and Sinks of Reactive Gases

Acknowledgments
NASA ROSES SEAC4RS
NASA ROSES ACCDAM
DC-8 pilots and crew
SEAC4RS science team

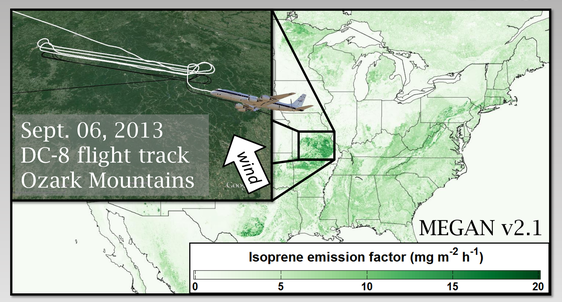
G. M. Wolfe[1,2], T. F. Hanisco[1], H. L. Arkinson[3], T. P. Bui[4], J. D. Crounse[5], J. Dean-Day[4,6], A. Goldstein[7], A. Guenther[8], S. R. Hall[9], G. Huey[10], D. J. Jacob[11], T. Karl[12], P. S. Kim[11], X. Liu[10], M. R. Marvin[3], T. Mikoviny[13], P. K. Misztal[7], T. B. Nguyen[5], J. Peischl[14,15], I. Pollack[14,15], T. Ryerson[14], J. M. St. Clair[5], A. Teng[5], K. R. Travis[11], K. Ullmann[9], P. O. Wennberg[5], and A. Wisthaler[13]
[1]NASA GSFC, [2]JCET/UMBC, [3]U. MD, [4]NASA ARC, [5]Caltech, [6]BAERI, [7]UC Berkeley, [8]PNNL, [9]NCAR, [10]GA Tech, [11]Harvard, [12]U. Innsbruck, [13]U. Oslo, [14]NOAA, [15]CIRES/CU Boulder

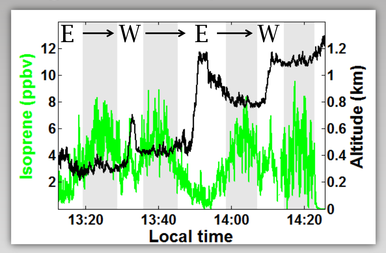
MOTIVATION

- Forests are both a source and sink of reactive gases
- Gaps in our understanding of **emissions**, **deposition** and **chemistry** collectively limit confidence in model predictions
- Disentangling processes with observations of chemical concentrations alone is challenging

MISSION

SEAC⁴RS: Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys



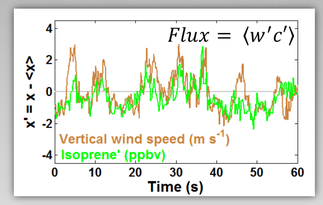


Gray regions denote "clean" background air

METHODS

Eddy Covariance

Concentrations vary in ↑ and ↓ turbulent eddies



Mass Balance

$$\frac{\partial C}{\partial t} = Q - \bar{U}\frac{\partial C}{\partial x} - \frac{\partial F}{\partial z}$$

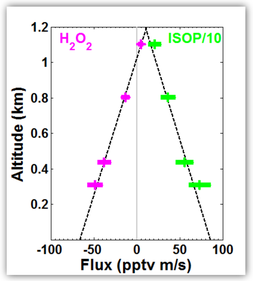
$$F(z) = \left(Q - \bar{U}\frac{\partial C}{\partial x} - \frac{\partial C}{\partial t}\right) z + F_0$$

flux — chemistry — advection — storage — emission / deposition

Fluxes provide direct constraints on the **rates** of physical and chemical processes.

RADICAL CYCLING

- Concentrations of OH and HO_2 derived from slope of isoprene and H_2O_2 flux vertical profiles
- Comparison with GEOS-Chem and UWCM shows good agreement for HO_2 but not OH



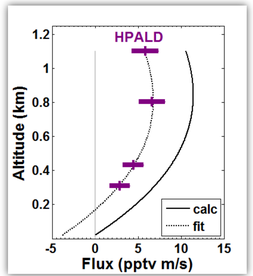
Fluxes provide an additional check on radical concentrations in efforts to improve model mechanisms and investigate measurement artifacts.

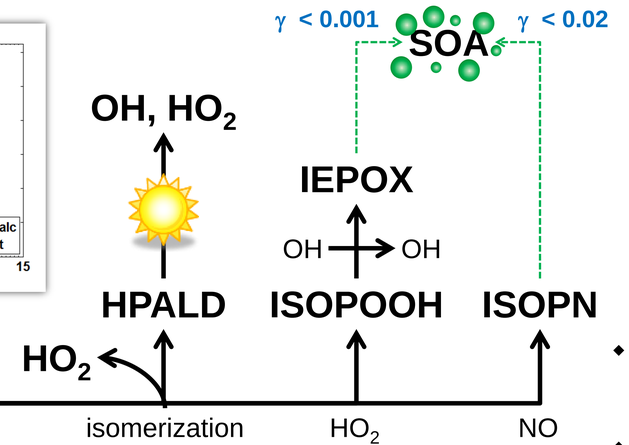
Source	[OH] 10^6 cm^{-3}	[HO_2] 10^8 cm^{-3}
Flux	1.3 ± 0.3	5.8 ± 1.0
GEOS-Chem	0.5 ± 0.1	5.8 ± 0.4
UWCM (0-D)	1.0 ± 0.4	6.6 ± 0.6

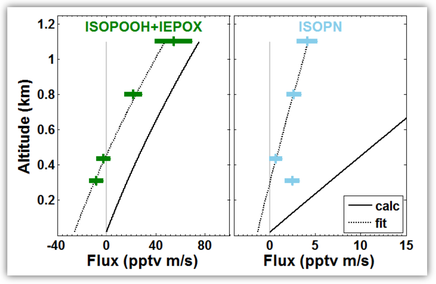
ISOMERIZATION

- Curvature reflects temperature dependence of isomerization
- Calculated flux profile using lab-derived HPALD production rate agrees with observed slope

Isomerization may be a less important radical source in low NO_x regimes than initially proposed.



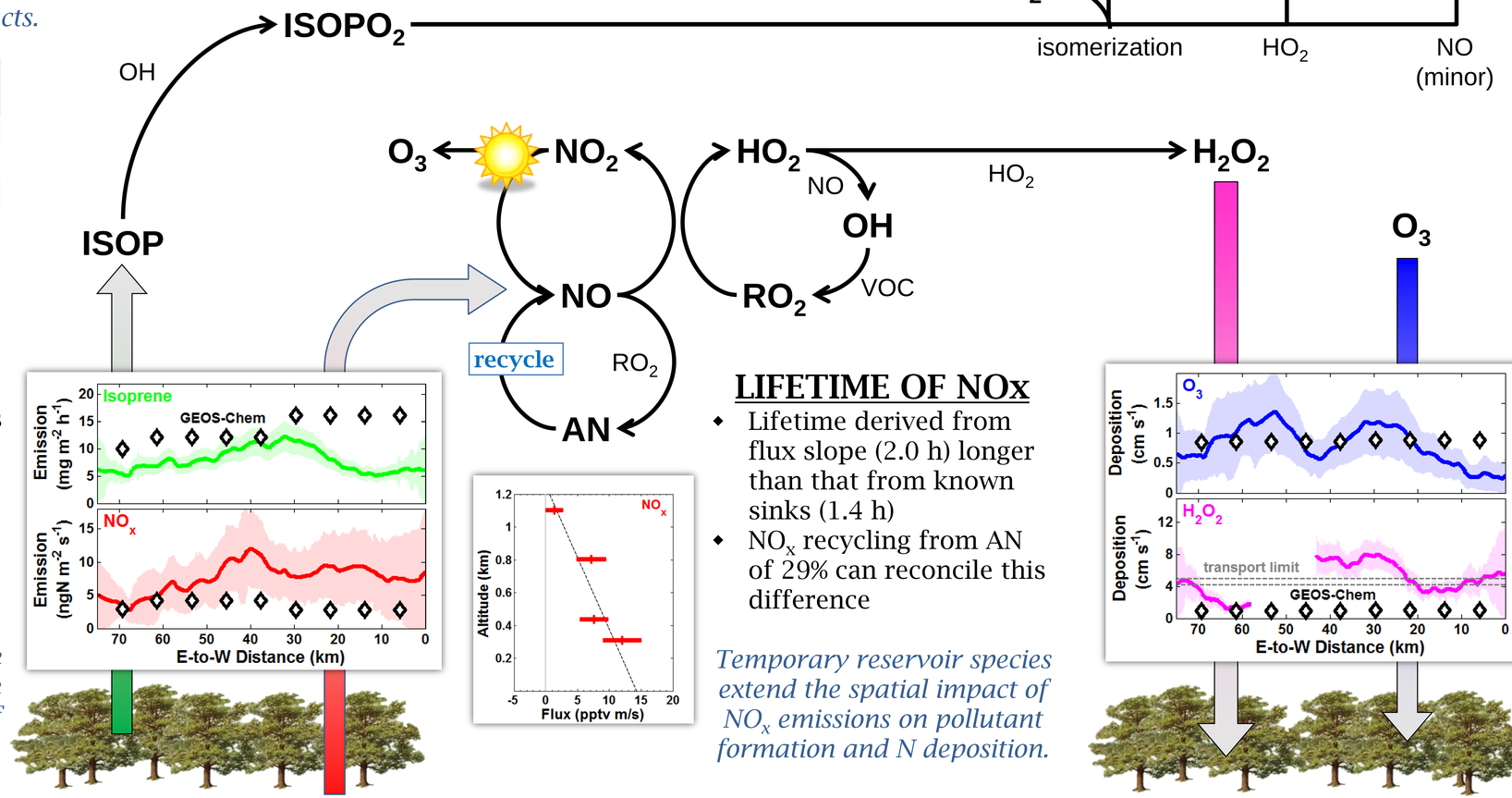




AEROSOL UPTAKE

- Difference in calculated vs observed slopes imply unknown sources/sinks
- ISOPOOH+IEPOX budget closed with minimal aerosol uptake
- 70% of ISOPN sink may be due to aerosol losses

Isoprene nitrates may be a source of particle mass, while IEPOX uptake may be inhibited (in this environment).



EMISSIONS

- Observed isoprene emissions are 40% lower than model
- Observed soil NO_x emissions are 50% higher than model
- Wavelet transforms illustrate how surface fluxes vary across the transect

Direct measurements of surface fluxes at an ecosystem scale are ideal for targeted refinement of emission inventories.

LIFETIME OF NOx

- Lifetime derived from flux slope (2.0 h) longer than that from known sinks (1.4 h)
- NO_x recycling from AN of 29% can reconcile this difference

Temporary reservoir species extend the spatial impact of NO_x emissions on pollutant formation and N deposition.

DEPOSITION

- Model accurately predicts O_3 deposition (for this case)
- H_2O_2 deposition consistent with transport-limited uptake
- Variability along transect may reflect both surface characteristics and chemistry

Parameterizations must be retooled to robustly reflect physical and chemical mechanisms driving deposition.